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Interactive comment on "Conceptual study on nucleation burst evolution in the convective boundary layer – Part I: Modelling approach" *by* O. Hellmuth

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Response to referee 2 (Michael Boy)

1. General remarks and change of title

See my response to referee 1 and 3.

2. Authors comment (AC) to specific comments of referee 2 (RC2)

2.1 RC2: Page 11418, line 23: Anticorrelation of sulphur dioxide and NPF according to Boy et al. (2003c)

AC: Page 11418, line 26: The following sentences have to be added:

"In forest regions Boy et al. (2003c) found, that SO_2 is not correlated, or even anticorrelated with the number concentration of small particles. As SO_2 is mainly of anthropogenic origin, it is mostly associated with high number concentrations of pre-existing aerosols, represententing polluted conditions. Previous findings from laboratory studies revealed a direct correlation between increasing SO_2 concentrations and measured number concentrations of newly formed particles in a ozone/ α-pinene reaction system (Boy et al., 2003c)."

See response to part IV.

The paper of Boy et al (2003c) deserves a detailed evaluation with respect to the interpretation of the present results. In response to RC2, I decided to discuss this paper in more detail in a technical note to be submitted to ACPD.

2.2 RC2: Page 11424, line 9: Recommendation to delay a sentence

AC: The incriminated half sentence

"... the detailed chemistry of the events remains uncertain ..."

directly refers to the citations, that follow there. However, the considered statement was already explained a few sentence earlier, hence, it is redundant. Deleted.

2.3 RC2: Page 11423, chapter 3.2.4: Missing of some interesting contributions concerning ion-induced nucleation. Would be a valuable part in this chapter.

AC: A new paragraph is inserted on page 11245, after line 14. See final version.

"Recently, the so-called ion-mediated nucleation has attracted growing attention, because atmospheric gaseous ions were found to can effectively induce NPF in two steps: (Eichkorn et al., 2002):

1. Ion-induced nucleation: (i) formation of a stable neutral molecular cluster; (ii) attachment of low volatility trace gas molecules to small ions leading to large cluster ions; (iii) ion-ion recombination of positive and negative cluster ions leading to an

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electrically neutral molecular cluster which may be sufficiently large to experience spontaneous growth by condensation of low volatility trace gas molecules;

2. Subsequent growth of the freshly nucleated cluster to a small volatile aerosol particles of detectable size $(D_p \geq 3\mathrm{nm})$.

There are two major effects of the net electric charge of an ion: (a) large stabilisation of a cluster ion compared to an electrically neutral cluster of the same size and composition; (b) much faster growth of a cluster ion due to the large ion-molecule collision cross section resulting from the long range of the charge-dipole interaction.
Favoured conditions for the occurrence of ion-induced nucleation are low temperatures and large concentrations of low volatility gases. The upper troposphere is suspected to be a favoured place for ion-mediated nucleation under certain conditions (Eichkorn et al., 2002; Lovejoy et al., 2004). A strong argument suggesting ion-mediated aerosol formation also results from the fact, that in both the homogeneous binary and ternary nucleation, the growth rate of newly-formed nano-particles from condensation of $\mathrm{H_2O}$, $\mathrm{H_2SO_4}$, and $\mathrm{NH_3}$ was found to be often much to low to explain the rapid appearance of fresh ultrafine aerosols during midday (Weber et al., 1997; Yu et al., 2000). Consequently, Yu et al. (2000) hypothesised that some of present day discrepancies between theory and observation may be due to the intervention of background ionisation in NPF. For further information, the reader is referred to, e.g., (*here a list of 13 references follows*) A detailed evaluation of recent finding on ion-mediated nucleation (level of understanding, comparison with classical nucleation theory, condition of occurrence, elementary processes, modelling approach, empirical findings) with respect to the present modelling approach will be given in a technical note, which is in preparation for Atmos. Chem. Phys. Discuss."

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 11413, 2005.